

PATENT ABSTRACTS OF JAPAN

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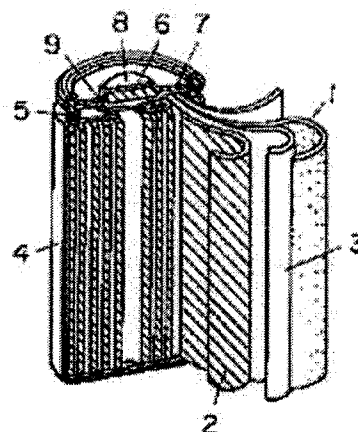
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(54) NEGATIVE ELECTRODE ACTIVE MATERIAL FOR ALKALINE STORAGE BATTERY AND BATTERY USING THIS

(57)Abstract:

PROBLEM TO BE SOLVED: To form a storage battery of an electrode of a small and thin type by using a compound having a quinone structure to form a complex with a metallic ion for an alkaline storage battery as an active material.

SOLUTION: In a negative electrode plate 1, 2,5-dichloro-3,6 dihydroxy-p- benzoquinone is agitated by an alkaline aqueous solution, for example, KOH, and after it is left as it is for a day, an obtained precipitate is filtered and dried, and a complex with a metallic cation is formed as an active material. This is mixed with carbon of a conductive material, a 1 wt.% aqueous solution of carboxymethyl cellulose is added and paste is made, a fluoro-resin is added as a binding agent, and is applied to both surfaces of a copper current collecting body, and is pressed to a prescribed thickness after drying, and a negative electrode is obtained. In a positive electrode plate 2, a conductive material and a nickel hydroxide are mixed, the 1 wt.% aqueous solution of carboxymethyl cellulose is added and paste is made, a fluoro-resin is added as a binding agent, and is applied to both surfaces of a nickel current collecting body, and is pressed to a prescribed thickness after drying, and a positive electrode is formed. Nonwoven fabric is used as a separator, and KOH is used as an electrolyte.



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CLAIMS

[Claim(s)]

[Claim 1]Negative electrode active material for alkaline batteries being a compound which has quinone structure and forms a metallic cation (Li^+ , Na^+ , K^+ , Ba^{2+}) and a complex.

[Claim 2]The negative electrode active material for alkaline batteries according to claim 1, wherein a compound has a hydroxyl ($-\text{OH}$) basis, a cyano ($-\text{CN}$) basis, or at least one or more halogen ($-\text{F}$, $-\text{Cl}$, $-\text{Br}$) bases in a functional group.

[Claim 3]An alkaline battery provided with a negative electrode using the active material according to claim 1, an anode using a metallic oxide, and an alkali electrolyte.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to the cell which used the negative electrode active material for alkaline batteries, and this.

[0002]

[Description of the Prior Art]In recent years, the portable device is strengthening the miniaturization tendency and the miniaturization of the storage battery which is the power supply inevitably, and high-energy-density-ization are desired.

[0003]Although the nickel cadmium battery which the storage battery using an alkali electrolyte used nickel hydroxide for positive active material, and used cadmium for negative electrode active material was used more widely than before, It worried about influence on the environmental problem of cadmium, and the nickel hydrogen storage battery using the hydrogen storing metal alloy as the alternative material has been put in practical use. This was able to attain the high capacity and densification which are simultaneously demanded of the storage battery.

[0004]Although it has the feature that charge and discharge are possible for an alkaline battery at a high current, and a cycle life is long, In recent years, in connection with portable one of various kinds of electronic equipment, and cordless making, development of the lithium ion battery which has the weight energy density beyond it is progressed and required to the weight energy density of nickel cadmium and a nickel hydrogen storage battery being about 70Whs/kg. However, in safety, it holds expectations of further small size and weight saving also for the alkaline battery which is dominance, and development is hurried.

[0005]The storage battery which used cadmium or a hydrogen storing metal alloy for negative electrode active material among the storage batteries using an alkali electrolyte has the large weight per unit volume of these active materials itself, and it is difficult to constitute small size and a thin storage battery. Therefore, material with a moldability small the weight per unit volume of the active material itself and sufficient was called for.

[0006]

[Problem(s) to be Solved by the Invention]The organic compound which has quinone structure is mentioned as one of the materials which were mentioned above. Using for a side chain the oxidation reduction polymer which has quinone structure as an electrode material in an acid aqueous solution and a nonaqueous solution as a thing using such a compound that has quinone structure as an active material (JP,H6-56989,A) is proposed.

[0007]However, although it succeeded in the trial which many compounds with quinone structure dissolve to an alkaline aqueous solution, is thin-film-ized as proposed in the above-mentioned gazette, and is used as a polymer battery, the application to the active material for alkaline batteries was very difficult.

[0008]This invention solves an aforementioned problem, and an object of this invention is to provide

an alkaline battery with small weight energy density with small size and a thin shape.

[0009]

[Means for Solving the Problem] This invention uses a compound which forms quinone structure **** and a metallic cation, and a complex as negative electrode active material to achieve the above objects.

[0010]

[A gestalt of the actual condition of an invention] Structure of 2,5-dichloro-3,6-dihydroxy-p-benzoquinone is shown in left-hand side of drawing 1 as a quinone compound. Although it was known that it is the reversible system of reaction, since it was meltable to an alkaline aqueous solution, p-benzoquinone was not able to be used as an electrode material of an alkaline battery.

[0011] However, by having hydroxyl in a position of the 2 or 5th place of benzoquinone structure like chloride which is a halogen group, and a compound which has hydroxyl in a position of the 3 or 6th place, If an alkaline aqueous solution is touched as shown in drawing 1, a metallic cation and a stable complex can be formed and the following electrochemical reaction will become possible. That is, if potential is swept in the direction of electrochemical reduction, while a proton will invade into a complex, a carbon-oxygen double bond of the para position is solved and OH combination is formed. Conversely, if potential is swept in the direction of electrochemical oxidation after a reduction reaction, OH combination of the para position will cause dehydrogenation and will form a carbon-oxygen double bond again.

[0012] Since the nucleophilic displacement of the chloride to which such an effect exists [in / like tetrachloro-p-benzoquinone / in a functional group / a halogen group] at least in one opposite is easy to be carried out, it forms the same stable complex as the above. Since a cyano group or a halogen group is the strong electronegativity which they have, it is effective in helping stabilization of ionicity combination with a metallic cation required for complexing.

[0013]

[Example] Next, the example of this invention is explained. Drawing 2 shows the nickel quinone storage battery by one working example of this invention. the negative electrode plate for which 1 used the quinone complex by this invention for the main ingredients, the anode board for which 2 used nickel hydroxide for the main ingredients, and 3 -- as for a safety valve and 7, a case and 5 are [a positive pole terminal and 9] positive electrode leads an obturation board and 8 an electric insulating plate and 6 a separator and 4.

[0014] In working example of this invention, the manufacturing method of the negative electrode plate 1 is shown. First, after ****(ing) 2,5-dichloro-3,6-dihydroxy-p-benzoquinone in the alkaline aqueous solution (KOH) and carrying out day neglect, the obtained sediment was filtered and dried and the complex with a metallic cation was obtained. This complex was mixed with the carbon which is a conducting material, the 1-% of the weight solution of carboxymethyl cellulose was added, the paste was built, fluororesin was further added as a binder, and copper charge collector both sides were plastered. After drying this, it pressed in predetermined thickness and was considered as the negative electrode.

[0015] Nickel hydroxide was mixed with the conducting material to the anode board 2, the 1-% of the weight solution of carboxymethyl cellulose was added to it, the paste was built, fluororesin was further added as a binder, and nickel charge collector both sides were plastered. After drying this, it pressed in predetermined thickness and was considered as the anode. In the separator, the electrode group was constituted using the nonwoven fabric made from polypropylene which performed hydrophilic processing, after inserting in metal casing and pouring in the caustic potash solution of specific gravity 1.3 as an electrolysis solution, it obturated and the nickel quinone storage battery was constituted.

[0016] The charge-and-discharge curve of this cell is shown in drawing 3. Battery sizes are AAA and nominal capacity 300mAh, and both charge and discharge currents were performed by 0.1C. Charge voltages were about 1.3 v and discharge voltage was about 1.15v.

[0017]In this working example, although the metallic cation which forms a complex by changing the solute of the alkaline aqueous solution which carries out day neglect could be changed, when metallic cations differed (lithium, sodium, barium), the same characteristic as the case of potassium was obtained.

[0018]It is thought that the same characteristic is obtained from such a mechanism also in the high molecular compound which has a complex which has quinone structure in a main chain.

[0019]It is thought by optimizing the manufacturing conditions of an electrode substrate and a negative electrode that still bigger current can be sent.

[0020]

[Effect of the Invention]According to this invention, the compound which has the quinone structure which forms a metallic cation and a complex can be used as an active material as mentioned above. Small size, a thin electrode, and a storage battery can be provided by using this active material for an alkaline battery.

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TECHNICAL FIELD

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PRIOR ART

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EFFECT OF THE INVENTION

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1]The mimetic diagram showing the structure of the compound which has the quinone structure used for one working example of this invention, and its metal complex

[Drawing 2]The outline lineblock diagram of the nickel quinone storage battery in one working example of this invention

[Drawing 3]The figure showing the charging and discharging characteristic of this example

[Description of Notations]

- 1 The negative electrode using a quinone compound
- 2 Nickel hydroxide anode
- 3 Separator

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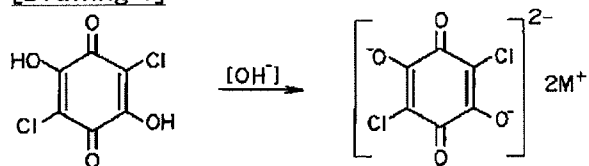
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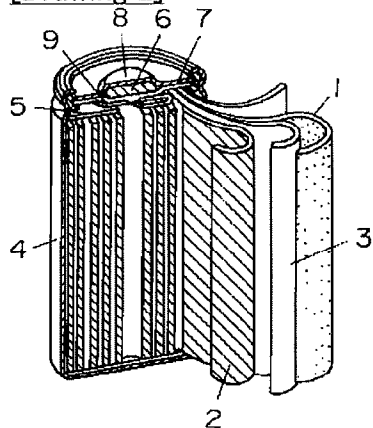
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DRAWINGS

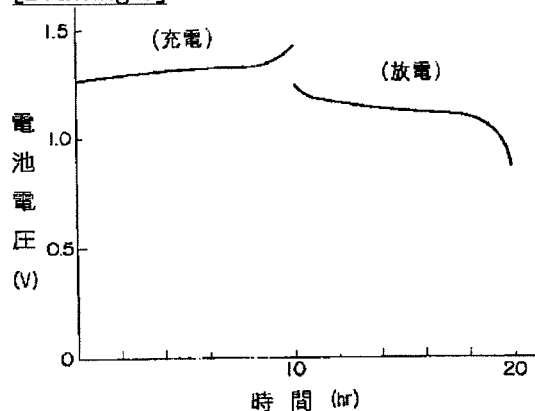
[Drawing 1]



[Drawing 2]



[Drawing 3]



[Translation done.]

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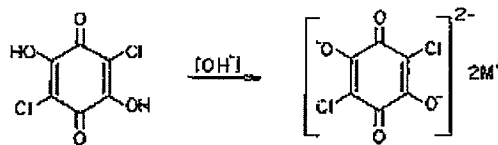
(74) 代理人 弁理士 池本 智之 (外1名)

(54) 【発明の名称】 アルカリ蓄電池用負極活物質とこれを用いた電池

(57) 【要約】

【課題】 小型・薄型化が可能で成形性の良いアルカリ蓄電池用負極活物質とこれを用いたアルカリ蓄電池を提供する。

【解決手段】 キノン構造を有する化合物を金属錯体としてアルカリ溶液中で安定化させ、負極活物質とし、この負極を用いてアルカリ蓄電池を構成する。



(2)

特開平10-294107

1

2

【特許請求の範囲】

【請求項1】 キノン構造を有し、金属陽イオン（Li⁺、Na⁺、K⁺、Ba²⁺）と錯体を形成する化合物であることを特徴とするアルカリ蓄電池用負極活物質。

【請求項2】 化合物は、官能基にヒドロキシル（-OH）基、シアノ（-CN）基あるいはハロゲン（-F、-Cl、-Br）基を少なくとも1つ以上有することを特徴とする請求項1記載のアルカリ蓄電池用負極活物質。

【請求項3】 請求項1記載の活物質を用いた負極と、金属酸化物を用いた正極と、アルカリ電解液とを備えたことを特徴とするアルカリ蓄電池。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、アルカリ蓄電池用の負極活物質およびこれを用いた電池に関するものである。

【0002】

【従来の技術】近年、ポータブル機器は小型化傾向を強めており、必然的にその電源である蓄電池の小型化、高エネルギー密度化が望まれている。

【0003】アルカリ電解液を用いた蓄電池は正極活物質に水酸化ニッケルを用い、負極活物質にカドミウムを用いたニッケル・カドミウム蓄電池が従来より広く使用されていたが、カドミウムの環境問題に対する影響が心配され、その代替物質としての水素吸蔵合金を用いたニッケル・水素蓄電池が実用化されてきた。これは同時に蓄電池に要求されている高容量、高密度化を図ることができた。

【0004】アルカリ蓄電池は、大電流で充放電が可能でサイクル寿命が長い特徴を有しているが、近年、各種の電子機器のポータブル、コードレス化に伴い、ニッケル・カドミウムおよびニッケル・水素蓄電池の重量エネルギー密度がおよそ70Wh/Kgであるのに対し、それ以上の重量エネルギー密度を有するリチウムイオン蓄電池の開発が進んできている。しかし、安全性において優位であるアルカリ蓄電池にもさなる小型・軽量化の期待がもたれ開発が急がれている。

【0005】アルカリ電解液を用いた蓄電池のうち、負極活物質にカドミウムあるいは水素吸蔵合金を用いた蓄電池は、これら活物質自体の単位体積当たりの重量が大きく、小型・薄型の蓄電池を構成することは困難である。そのため活物質自体の単位体積当たりの重量が小さく、成形性の良い材料が求められていた。

【0006】

【発明が解決しようとする課題】上述したような材料の一つとして、キノン構造を有する有機化合物が挙げられる。このような、キノン構造を有する化合物を活物質として用いたものとしては、酸水溶液および非水溶液中で側鎖にキノン構造を有する酸化還元ポリマーを電極材として用いること（特開平6-56989号公報）が提案

されている。

【0007】しかし、キノン構造を持つ多くの化合物はアルカリ水溶液に対して溶解してしまい、上記公報で提案されているように薄膜化して、ポリマー電池として使用する試みが為されてはいるものの、アルカリ蓄電池用の活物質への応用は大変困難であった。

【0008】本発明は上記課題を解決するものであり小型・薄型であって重量エネルギー密度が小さなアルカリ蓄電池を提供することを目的とする。

【0009】

【課題を解決するための手段】本発明は上記目的を達成するために、負極活物質としてキノン構造持ち、かつ金属陽イオンと錯体を形成する化合物を用いるものである。

【0010】

【発明の実施の形態】キノン化合物として2,5-ジクロロ-3,6-ジヒドロキシ-p-ベンゾキノンの構造を図1の左側に示す。p-ベンゾキノンは、可逆な反応系であることが知られているが、アルカリ水溶液に可溶であるためアルカリ蓄電池の電極材としては用いることができなかった。

【0011】しかしながら、ベンゾキノンの構造の2,5位の位置にハロゲン基であるクロライドと3,6位の位置にヒドロキシル基を有する化合物のようにヒドロキシル基を有することにより、図1に示すとおりアルカリ水溶液に触れると金属陽イオンと安定な錯体を形成することができ、次のような電気化学反応が可能になる。すなわち、電気化学的還元方向へ電位を掃引すると、プロトンが錯体に侵入するとともに、パラ位の炭素-酸素2重結合が解かれ、OH結合を形成する。逆に還元反応の後に電気化学的酸化方向へ電位を掃引すると、パラ位のOH結合は脱水素反応を起こし、炭素-酸素2重結合を再び形成する。

【0012】このような効果は、テトラクロロ-p-ベンゾキノンのように官能基がハロゲン基のみに於いても1つの対向位に存在するクロライドは求核置換されやすいため、上記と同様な安定な錯体を形成する。さらに、シアノ基あるいはハロゲン基は、それらが有する強い電気陰性度のため錯体形成に必要な金属陽イオンとのイオン性結合の安定化を助ける効果がある。

【0013】

【実施例】次に本発明の具体例を説明する。図2は本発明の一実施例によるニッケル・キノン蓄電池を示す。1は本発明によるキノン錯体を主成分に用いた負極板、2は水酸化ニッケルを主成分に用いた正極板、3はセパレータ、4はケース、5は絶縁板、6は安全弁、7は封口板、8は正極端子、9は正極リードである。

【0014】本発明の実施例において負極板1の作製方法について示す。まず、2,5-ジクロロ-3,6-ジヒドロキシ-p-ベンゾキノンをアルカリ水溶液中（KOH）で攪拌

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し、一日放置した後、得られた沈殿物をろ過、乾燥させ金属陽イオンとの錯体を得た。この錯体を導電材であるカーボンと混合し、カルボキシメチルセルロースの1重量%水溶液を加えてペーストをつくり、さらに結着剤としてフッ素系樹脂を加え銅集電体両面に塗着した。これを乾燥後、所定の厚みにプレスして負極とした。

【0015】正極板2には、導電材と水酸化ニッケルを混合し、カルボキシメチルセルロースの1重量%水溶液を加えてペーストをつくり、さらに結着剤としてフッ素系樹脂を加えニッケル集電体両面に塗着した。これを乾燥後、所定の厚みにプレスして正極とした。セパレータには親水処理を施したポリプロピレン製の不織布を用いて電極群を構成して、金属ケースに挿入して電解液として比重1.3の苛性カリ水溶液を注液した後、封口しニッケル・キノン蓄電池を構成した。

【0016】図3にこの電池の充放電曲線を示す。電池サイズはAAA、公称容量300mAhであり、充放電電流はともに0.1Cで行った。充電電圧は約1.3V、放電電圧は約1.15Vであった。

【0017】この実施例において、一日放置するアルカリ水溶液の溶質を変えることにより錯体を形成する金属陽イオンを変えることができるが、金属陽イオンが異なる場合（リチウム、ナトリウム、バリウム）においても*

*カリウムの場合と同様な特性が得られた。

【0018】なお、このようなメカニズムからキノン構造を有する錯体を主鎖にもつ高分子化合物においても同様な特性が得られるものと考えられる。

【0019】さらに、電極基板および負極の作製条件を最適化することにより、さらに大きな電流を流すことができると思われる。

【0020】

【発明の効果】以上のように本発明によれば、金属陽イオンと錯体を形成するキノン構造を有する化合物を活性物質として用いることができる。また、この活性物質をアルカリ蓄電池に用いることにより小型・薄型の電極および蓄電池を提供できる。

【図面の簡単な説明】

【図1】本発明の一実施例に用いたキノン構造を有する化合物とその金属錯体の構造を表す模式図

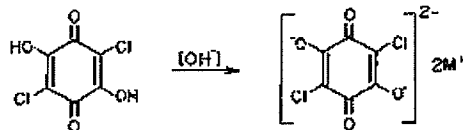
【図2】本発明の一実施例におけるニッケル・キノン蓄電池の概略構成図

【図3】本実施例の充放電特性を示す図

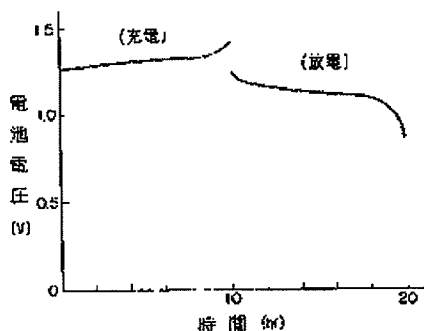
【符号の説明】

- 1 キノン化合物を用いた負極
- 2 水酸化ニッケル正極
- 3 セパレータ

【図1】



【図3】



【図2】

